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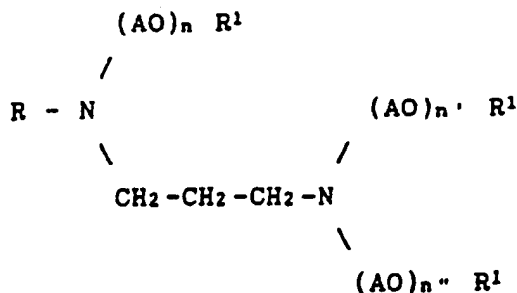
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(54) Concentrated herbicidal compositions based on N-phosphonomethylglycine

(57) Liquid herbicidal compositions which consist of aqueous solutions containing:

- a) N-phosphonomethylglycine (glyphosate; $(\text{HO})_2\text{P}(\text{O})\text{CH}_2\text{NHCH}_2\text{COOH}$) and/or one of its derivatives in a proportion of at least 40 g/l of glyphosate equivalent, and
b) a surfactant of formula



[in which

R is a C_{8-22} linear or branched, alkyl or alkenyl chain;

A denotes an alkylene group;

n, n' and n'' are integers such that $n + n' + n''$ is between 1 and 15; and

R¹ is hydrogen or an acyl radical] which has the nature of a biological activator/herbicide.

Incorporation of the above surfactant enables concentrated glyphosate compositions to be formulated.

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HERBICIDAL COMPOSITIONS BASED ON N-PHOSPHONOMETHYLGLYCINE

The present invention relates to new concentrated formulations based on N-phosphonomethylglycine or on compounds containing the N-phosphonomethylglycyl group.

5 N-Phosphonomethylglycine (sometimes called glyphosate), together with analogous compounds, their herbicidal properties and the formulations which contain them, are described in particular in US Patent 3,799,758. Although many glyphosate derivatives which
10 are water-soluble or insoluble are known, it is in fact very generally preferred to employ the water-soluble derivatives, and this is why it is the N-phosphonomethylglycine salts which have been generally developed or commercialized, especially the
15 isopropylammonium salt.

 More recently (European Patent Application No. 290,416), attempts have been made to develop concentrates based on N-phosphonomethylglycine salts, which may contain N-phosphonomethylglycine in acidic
20 form, but, in any event, containing this N-phosphonomethylglycine and/or its derivatives in soluble or solubilized forms, the feature of these concentrates being the presence of an alkoxyated amine of a particular type. This alkoxyated amine must
25 contain not more than 12 alkoxy groups per molecule and it must have the nature of a surface-active agent and it must promote the herbicidal activity of the

N-phosphonomethylglycine derivatives. It can be employed in a smaller quantity than the known surfactants of the known N-phosphonomethylglycine formulations, at least with regard to the production of concentrates intended to be applied in the open air, in the form of dilute slurries, at a rate of 100 to 600 l/ha. When the N-phosphonomethylglycine-based compositions are compositions containing an ammonium salt such as ammonium sulphate, the ethoxylated amine used in this Patent 290,416 is, in fact, an ethoxylated amine containing only 2 ethoxy groups per molecule.

An objective of the present invention is to provide concentrated compositions or formulations (also called concentrates) based on N-phosphonomethylglycine, and especially based on its water-soluble salts.

Another objective of the present invention is to provide concentrates based on N-phosphonomethylglycine or on its derivatives giving rise to dilute spraying mixtures which have a good herbicidal activity.

Another objective of the invention is to provide concentrates based on N-phosphonomethylglycine salts using new types of surface-active agents promoting the biological activity.

Another objective of the invention is to provide concentrates based on N-phosphonomethylglycine salts using surfactants which have a capability for

improved solubilization.

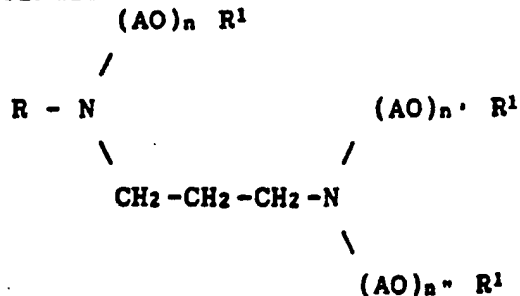
Another objective of the invention is to provide concentrates based on N-phosphonomethylglycine salts using new types of surface-active agents which are compatible in concentrated solutions with certain inorganic salts and especially with ammonium sulphate.

It has now been found that these objectives could be wholly or partially attained by virtue of the compositions according to the invention. In what follows the percentages are percentages by weight unless specifically stated. Furthermore, glyphosate equivalent is the name given to the corresponding quantity of product if all the N-phosphonomethylglycine derivative was in the form of ordinary N-phosphonomethylglycine.

These compositions are liquid compositions consisting of aqueous solutions containing:

a) N-phosphonomethylglycine and/or one of its derivatives in a proportion of at least 40 g/l of glyphosate equivalent,

b) a surfactant (having the nature of an activator) of formula



in which

R is a linear or branched, alkyl or alkenyl chain containing from 8 to 22 carbon atoms

A denotes an alkylene group, preferably
5 ethylene or propylene

n, n' and n" are integers such that $n + n' + n''$ is between 1 and 15, preferably 3 and 12

R¹ is the hydrogen atom or an acyl radical such as formyl, acetyl or propanoyl (that is to say
10 $\text{CH}_3\text{-CH}_2\text{-CO}$).

Naturally, this single surfactant may be replaced by a mixture of surfactants in which, on average, R, n, n' and n" correspond to the definitions given above.

15 An N-phosphonomethylglycine derivative means a compound containing the chain sequence $\text{-CO-CH}_2\text{-N-CH}_2\text{-P=O}$ (the nitrogen atom having a free valency and the phosphorus atom having two free valencies), preferably a salt, ester or amide, these terms being taken in the
20 broad sense, for example so as to include the sulphonamides.

The present invention advantageously relates to the concentrated compositions such as described above and additionally relating to one or other of the
25 following features:

a) the N-phosphonomethylglycine is in a quantity not exceeding the solubility limit in the medium in question, preferably between 60 and 200 g/l and still more preferably between 90 and 150 g/l,

5 b) the concentrated solution contains an ammonium (NH_4^+) salt such as the nitrate, phosphate, sulphamate, thiocyanate or preferably the sulphate, in a proportion of 50 to 400 g/l, preferably 100 to 300 g/l,

10 c) the concentrated compositions are intended to be diluted by the agriculturists in containers containing water so as to make it possible to spread these diluted spraying mixtures at a rate of 100 to 600 l/ha, the active substance, for its part, being applied at a rate of 0.125 to 4.5 kg/ha,

15 d) the weight ratio glyphosate equivalent/surfactant is between 0.6 and 6, preferably between 1 and 3,

 e) the solvent is water.

20 The relatively high quantity of surfactant shown above relates to this surfactant with the nature of a biological activator/herbicide, it being understood that the compositions according to the invention may additionally contain, as will be

explained later, all kinds of other components, and especially surfactants of the most diverse kinds and wetting in nature; these surfactants are then employed in dosages which are much lower than the activator
5 dosage.

The concentrated liquid compositions according to the invention are usually prepared merely by mixing the constituents.

The compositions according to the invention
10 usually contain from 5 to 25 % (preferably from 7 to 20 %) of active substances (herbicides), from 0.5 to 40 % (preferably from 10 to 20 %) of surface-active agent(s) of an activating nature, from 10 to 50 % (preferably from 20 to 30 %) of an ammonium salt
15 (adjuvant), from 0.1 to 10 % of surface-active agent(s) of a wetting nature, from 0 to 30 % of suitable additives such as foam suppressors, corrosion inhibitors, sequestrants, stabilizers, penetrating agents or adhesives. The compositions according to the
20 invention may, of course, also contain all the solid or liquid additives corresponding to the usual techniques of making up a formulation.

According to what has already been said, the concentrated compositions according to the invention
25 may contain one or more surface-active agents besides the surfactant with an activating nature defined above. The surface-active agent employed may be a wetting

agent of ionic or nonionic type or a mixture of such surface-active agents. There may be mentioned, for example, polycondensates of ethylene oxide with fatty alcohols or with fatty acids or with fatty amines, substituted phenols (especially alkylphenols or arylphenols), salts of esters of sulphosuccinic acids, taurine derivatives (especially alkyltaurates), phosphoric esters of ethylene oxide polycondensates with alcohols or phenols, esters of fatty acids and of polyols, and derivatives of the above compounds containing sulphate, sulphonate and phosphate functional groups.

In addition to N-phosphonomethylglycine and/or its derivatives, the compositions used in the invention may contain other known active substances with herbicidal properties or with plant growth-regulating properties.

As herbicidal substance which may be mixed with the glyphosphate derivatives in the compositions according to the invention there may be mentioned acifluorfen (or its sodium salt), aclonifen, bifenox, diflufenican, asulam, triazines (especially simazine and atrazine), diuron and oxadiazon, herbicides of hormone or phenoxy types, especially 2,4-D, [2,4-DB], MCPP, hydroxynonitriles (especially bromoxynil and ioxynil), imidazolinones (especially imazaquin and imazapur) and sulphonylureas (especially chlorsulfuron

and metsulfuron). The above names are standardized names used to denote the herbicides.

5 In most cases these herbicides are employed in a proportion of 1 to 400 parts by weight per 100 parts of glyphosate or of glyphosate equivalent. By employing the term glyphosate equivalent the calculation of the parts is thus reduced as if all the glyphosate derivatives were in the form of N-phosphonomethylglycine.

10 More precisely, when herbicidal active substances other than glyphosate are employed and are mixed with glyphosate, the following proportions are generally used for the weight ratio wr:

(wr is equal to

15 glyphosate or glyphosate equivalent/other herbicide)

glyphosate + acifluorfen:	$4/100 \leq wr \leq 9/100$
glyphosate + diuron or oxadiazon:	$2 \leq wr \leq 4$
glyphosate + aclonifen:	$1/3 \leq wr \leq 10/3$
glyphosate + bifenox:	$1/3 \leq wr \leq 1$
20 glyphosate + diflufenican:	$2 \leq wr \leq 20$
glyphosate + asulam:	$1/4 \leq wr \leq 1$
glyphosate + phenoxy:	$2/3 \leq wr \leq 4$
glyphosate + hydroxybenzonitrile:	$1 \leq wr \leq 10$
glyphosate + triazine:	$1/3 \leq wr \leq 1$
25 glyphosate + imidazolinone:	$1 \leq wr \leq 4$
glyphosate + sulph nylurea:	$100/6 \leq wr \leq 100$

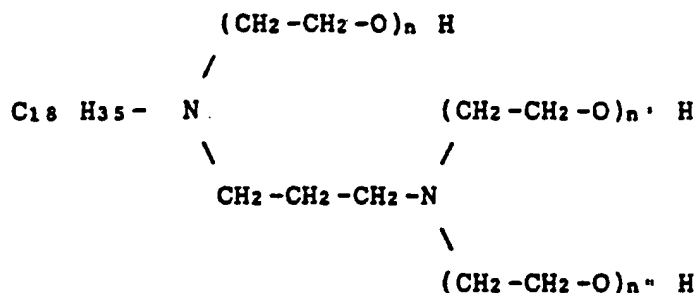
As a general rule, the herbicidal compositions used in the invention usually contain approximately from 0.05 to 95 % of one or more herbicidal active substances, approximately from 1 % to 95 % of one or more liquid carriers and optionally approximately from 0.1 to 50 % of one or more surface-active agents.

Examples of solutions according to the invention and examples of use of these solutions are given below by way of an example given without any limitation being implied. The active substance in these examples is N-phosphonomethylglycine in the form of isopropylammonium salt.

EXAMPLE 1

- N-Phosphonomethylglycine in the form of isopropylammonium salt 100 g/l
- Ammonium sulphate 200 g/l
- Surfactant of formula defined below 100 g/l
- Water sufficient quantity for 1 l

The surfactant used in this example was a product of formula



with $n + n' + n'' = 3$

EXAMPLE 2

- N-Phosphonomethylglycine in the form of isopropylammonium salt 100 g/l
- 5 - Ammonium sulphate 100 g/l
- Surfactant employed in Example 1 200 g/l
- Water sufficient quantity for 1 l

EXAMPLE 3

- N-Phosphonomethylglycine in the form of isopropylammonium salt 100 g/l
- 10 - Ammonium sulphate 150 g/l
- Surfactant employed in Example 1 150 g/l
- Water sufficient quantity for 1 l

EXAMPLE 4

- 15 - N-Phosphonomethylglycine in the form of is propylammonium salt 100 g/l
- Ammonium sulphat 300 g/l

- Surfactant employed in Example 1 100 g/l
- Water sufficient quantity for 1 l

EXAMPLE 5

- N-Phosphonomethylglycine in the form
5 of isopropylammonium salt 100 g/l
- Acifluorfen in the form of sodium salt 4.5 g/l
- Ammonium sulphate 200 g/l
- Surfactant of the formula defined
above with $n + n' + n'' = 10$
- 10 - Water sufficient quantity for 1 l

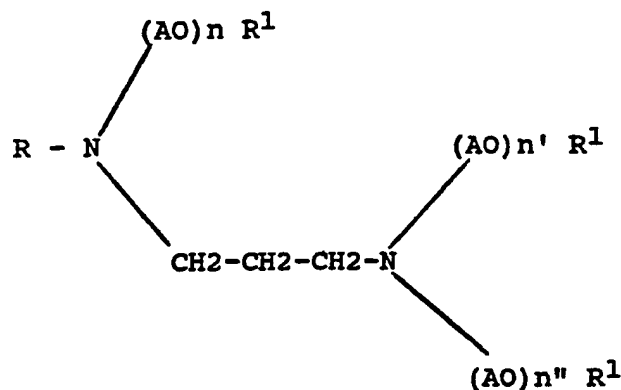
These solutions are diluted in water in a proportion of 2.5 and 5 l in 300 l and the spraying mixtures thus obtained are sprayed onto various adventitious plants.

- 15 The compositions of Examples 1 to 4, employed in the dosage of 250 g/l of active substance calculated in the form of N-phosphonomethylglycine, exhibit an effectiveness on various weeds, especially morning glory, purslane and Indian mallow, which is markedly
- 20 superior to that of commercial products, and especially of the product marketed under the trademark Round-up and which contains the same active substance in combination with ordinary surface-active agents.

CLAIMS

1. A liquid herbicidal composition which comprises an aqueous solution containing:

- a) N-phosphonomethylglycine and/or one of its derivatives in a proportion of at least 40 g/l of glyphosate equivalent, and
b) a surfactant of formula



in which

R is a linear or branched, alkyl or alkenyl chain containing from 8 to 22 carbon atoms

A denotes an alkylene group,

n, n' and n'' are integers such that n + n' + n'' is from 1 to 15

Each R¹, which may be the same or different is hydrogen or an acyl radical.

2. The composition according to claim 1, wherein A is an ethylene or propylene group.

3. The composition according to claim 1 or 2,
wherein $n + n' + n''$ is between 3 and 12.

4. The composition according to any one of claims 1
to 3, wherein R^1 is a formyl, acetyl or propanoyl radical,
5 preferably acetyl.

5. The composition according to any one of claims 1
to 4, wherein the N-phosphonomethylglycine and/or one of
its derivatives is in a quantity not exceeding the
solubility limit in the medium in question and is from 60
10 to 200 g/l.

6. The composition according to any one of claims 1
to 5, wherein the N-phosphonomethylglycine and/or one of
its derivatives is in a quantity of from 90 to 150 g/l.

7. The composition according to any one of claims 1
15 to 6, wherein the concentrated solution contains an
ammonium salt such as the nitrate, phosphate, sulphamate,
thiocyanate or preferably the sulphate, in a proportion of
50 to 400 g/l.

8. The composition according to any one of claims 1
20 to 7, wherein the concentrated solution contains an
ammonium salt in a proportion of 100 to 300 g/l.

9. The composition according to any one of claims 1
to 8, which is intended to be diluted in water so as to
make it possible to spread these diluted spraying mixtures
25 at a rate of 100 to 600 l/ha, the active substance, for its
part, being applied at a rate of 0.125 to 4.5 kg/ha.

10. The composition according to one of claims 1 to 9, wherein the weight ratio glyphosate equivalent/surfactant is from 0.6 to 6.

11. The composition according to claim 10, wherein
5 the weight ratio is from 1 to 3.

12. The composition according to any one of claims 1 to 11, wherein the solvent is water.

13. The composition according to any one of claims 1 to 12, which comprises other surfactants of a
10 wetting nature and optionally one or more other additives selected from foam suppressors, corrosion inhibitors, sequestrants, stabilizers, penetrating agents or adhesives.

14. The composition according to any one of claims 1 to 13, which comprises from 5 to 25 % of
15 herbicidal active substance, from 0.5 to 40 % of surface-active agent(s) of an activating nature, from 10 to 50 % of an ammonium salt (adjuvant), from 0.1 to 10 % of surface-active agent(s) of a wetting nature, and from 0 to 30 % of other additives.

20 15. The composition according to any one of claims 1 to 14, which comprises from 7 to 20 % of herbicidal active substance, from 10 to 20 % of surface-active agent(s) of an activating nature, and from 20 to 30 % of ammonium salt.

25 16. The composition according to any one of claims 1 to 15, which comprises at least one other

herbicide in addition to N-phosphonomethylglycine and/or one of its derivatives.

17. The composition according to claim 16 wherein the other herbicide chosen from the group consisting of:
5 acifluorfen (or its sodium salt), aclonifen, bifenox, diflufenican, asulam, triazines (especially simazine and atrazine), diuron and oxadiazon, herbicides of hormone or phenoxy types, especially 2,4-D, [2,4-DB], MCPP, hydroxybenzonitriles (especially bromoxynil and ioxynil),
10 imidazolinones (especially imazaquin and imazapur) and sulphonylureas (especially chlorsulfuron and metsulfuron).

18. The composition according to claim 17, wherein the said other herbicides are present in the following weight ratio wr:

15 (wr is equal to
glyphosate or glyphosate equivalent/other herbicide)
glyphosate + acifluorfen: $4/100 \leq wr \leq 9/100$
glyphosate + diuron or oxadiazon: $2 \leq wr \leq 4$
glyphosate + aclonifen: $1/3 \leq wr \leq 10/3$
20 glyphosate + bifenox: $1/3 \leq wr \leq 1$
glyphosate + diflufenican: $2 \leq wr \leq 20$
glyphosate + asulam: $1/4 \leq wr \leq 1$
glyphosate + phenoxy: $2/3 \leq wr \leq 4$
glyphosate + hydroxybenzonitrile: $1 \leq wr \leq 10$
25 glyphosate + triazine: $1/3 \leq wr \leq 1$
glyphosat + imidazolinone: $1 \leq wr \leq 4$

glyphosate + sulphonylurea: $100/6 \leq wr \leq 100$.

19. The composition according to one of claims 16 to 18, which comprises acifluorfen or its sodium salt.

20. The composition according to claim 1
5 substantially as hereinbefore described.

21. A process for combating weeds, in which a composition according to one of claims 1 to 20 is employed.

22. The process according to claim 21, wherein the said composition is diluted and applied at a rate of 100 to
10 600 l/ha, the active substance being applied at a rate of 0.125 to 4.5 kg/ha.

23. The process according to claim 21 or 22 substantially as hereinbefore described.